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Colloid-polymer mixtures and the limits of criticality

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Since the industrial revolution, boiling and condensation have come to be one of the best recognised phase transitions of condensed matter. Approaching the critical point, a liquid becomes indistinguishable from its vapour, the interfacial thickness diverges and the system is entirely dominated by long-wavelength density fluctuations so microscopic properties such as chemical structure may be safely neglected. This universality concept assumes a large separation between the lengthscales of the critical density fluctuations and the constituent particles. However, the connection between these mesoscopic and microscopic regimes remains a long-standing open question. Here we access this “missing link” for the first time by using a mesoscopic analogue of simple liquids, colloid-polymer mixtures.

Direct visualization of both the individual colloidal particles and critical density fluctuations provides full access to all lengthscales. We reveal the first particle-level images of the critical clusters and liquid-gas interface. It turns out that the lengthscale of both is identical. Surprisingly, we find that the behaviour can be very well described by critical scaling almost to the single-particle level, as shown by the correlation length, critical cluster fractal dimension and free energy, all of which we directly measure. However, when the correlation length falls to 2-3 times the constituent particle size, we reveal a clear crossover from critical to non-critical behaviour. Our results provide a framework in which to unify the disparate particle- and correlation- lengthscales, and bring new insight into the nature of the liquid-gas interface and the limit of the critical regime.

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